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OAK RIDGE NATIONAL LABORATORY EVALUATION FOR DRUM CHARACTERIZATION AND SOURCE TERM

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OAK RIDGE NATIONAL LABORATORY
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LETTER REPORT

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DRUM CHARACTERIZATION INTEGRATED SUMMARY REPORT

1. DRUM HISTORY

1.1 Summary Conclusion

No evidence has been found to suggest Drum 68660 was processed, handled, transported, or stored any differently from other processed LANL MIN02 waste drums. A study of the physical and environmental effects this drum experienced during its lifetime was performed and documented to make this conclusion. Data were acquired from several sources, including Waste Characterization, Reduction, and Repackaging Facility (WCRRF) waste packaging reports, Real Time Radiographic (RTR) videos, Los Alamos spreadsheets prepared during their investigations, and conversations with multiple individuals. The timeline is being used to understand the sequence of events, and as input into the drum chemistry modeling efforts.

Also included was a determination of detailed inventory for the contents of the drum based on historical knowledge for the processes resulting the original MIN02 waste salts, WCRRF processing waste packages, post-processing RTRs, and Central Characterization Project (CCP) high efficiency passive neutron counter (HENC) used to perform non-destructive assays of the drums before placement into the WIPP repository. The drum inventory also supports the investigation into possible chemical reactions leading to the event.

1.2 Process History for Recovery of Plutonium

The MIN02 waste salts were produced during TA-55 evaporator operations which was used to minimize bulk volumes of anion exchange (AEX) effluents produced during the recovery of plutonium. Among the evaporator feed solutions relevant to this investigation generated from AEX effluent are the lean residue feed (LR), rich residue feed (RR), distillate solution feed (DS), or the filtrate from oxalate precipitation (OX) on the AEX plutonium effluent solution. The designator IXFS was used prior to LR/RR/DS to generally identify nitric acid ion exchange evaporator feed solutions. These designators were used as part of a naming convention to allow the evaporator feed that produced a certain bag of salt to be identified back to the process used to recover the plutonium. According to LANL documentation and procedures, 500-600 L of feed was reduced to 10-25 L of “bottoms”. [MST-12 Procedure 485-REC-R00 and 485-REC-R01]. The “bottoms” were cooled to room temperature which produces the nitrate salts and a liquid supernate. The liquid was separated from the salts by filtration using a 200 mesh stainless steel screen. After filtration, the salts were vacuum dried. This process consisted of pulling air through the salts using house vacuum for approximately 15 minutes. After 1985 procedures were modified to allow for washing of the salts to meet an Economic Discharge Limit (EDL). Although it was not consistently documented it was felt by LANL personnel that it was likely that the nitrate salts produced in the 1980s onward were washed with concentrated or 7 M nitric acid as needed to meet the EDL. Nitrate salts produced from oxalate precipitation were washed with water. It was cautioned in the procedure to do this to prevent “…decomposition of any oxalic acid present in the salts and could result in pressurization of the sealed 55-gallon drum containing the salts.” The “dried” salts were placed into a plastic bag and a grab sample, possibly from multiple places within the bag, was taken for americium and plutonium analysis prior to the bag being sealed. The sealed plastic bag was then double bagged, bagged out of the glovebox, and placed into a waste drum. These waste drums are the parents of the processed daughter drums produced during the recent campaign at LANL, a sub-set of which were placed in WIPP Panel 7 for permanent storage.

From the designators for the evaporator feeds general assumptions as to the make-up of the salts can be made.

First, when plutonium material is collected using anion exchange the liquid effluent contains most of the americium. Therefore when the liquid effluent is run through the evaporators it produces salts with high Am/SNM (Pu239 and Pu240) ratios. These are the LR and RR items from 1985 and forward. Before 1985 they are the IXFS items. When the Pu on the columns is removed and precipitated with oxalic acid the resultant liquids filtered off of the oxalate precipitates are very low in americium and the salts produced from evaporation have a low Am/SNM ratios. These salts have the OX or UOX designator in their sample identifiers. This information as well as historical analytical data for the salts was used for the evaluation for source term.

Secondly, typically 10 – 20% oxalic acid was used for the oxalate precipitation process. The excess oxalic acid remains in the solution sent to the evaporators. LANL analysis of the OX and UOX salts suggests that the majority of the oxalate does not precipitate upon evaporation. This means most of the oxalate remains in the post-evaporator liquid that was filtered off for processing by cementation. However, the salt's interstitial liquids would contain oxalate. Therefore, the amount of oxalate remaining with the nitrate salts depends upon how much liquid remains with the precipitated salts. This is considered to be a low weight percent of the total salt mass. This information assisted in the effort to identify and model possible chemical reactions during this investigation.

Thirdly, 1995 LANL characterization of LR, OX, and DS evaporator feed bottoms provided information as to the general chemical makeup of these salt types for use in the study of models for chemical reactivity.

1.3 Drum History

Drum 68660 was produced as part of the processing of parent drum S855793 (identified as drum S855793 in the WCRRF package). Radiography indicated the presence of liquid in the 55 gallon drum leading to the required processing. Two sibling drums were produced during processing, 68660 and 68685. Drum 68660 was shipped to WIPP. Drum 68685 remains at Los Alamos.

Figure E-1 depicts the processing and indicates the materials placed in each of the sibling drums. Drum 68660 received all of the observed 2 gallons of processed liquid (nitric acid, TEA, Swheat Scoop®), one process facility tungsten/bismuth/lanthanum-impregnated glovebox glove, job control solid waste (empty TEA bottles), and some of the processed nitrate/oxalate salts. The material is in three layers, and the drum is ~60% full. It should be noted that the WCRRF processing package for this parent drum recorded that glovebox gloves (plural) were added to 68660; however, a single glove was visually identified by an expert RTR operator at INL during review of the original RTR (See Addendum A to this appendix).

Drum 68685 received the remainder of the processed nitrate/oxalate salts (salts and Swheat Scoop®) as well as the lead blanket. It is approximately 85% full.

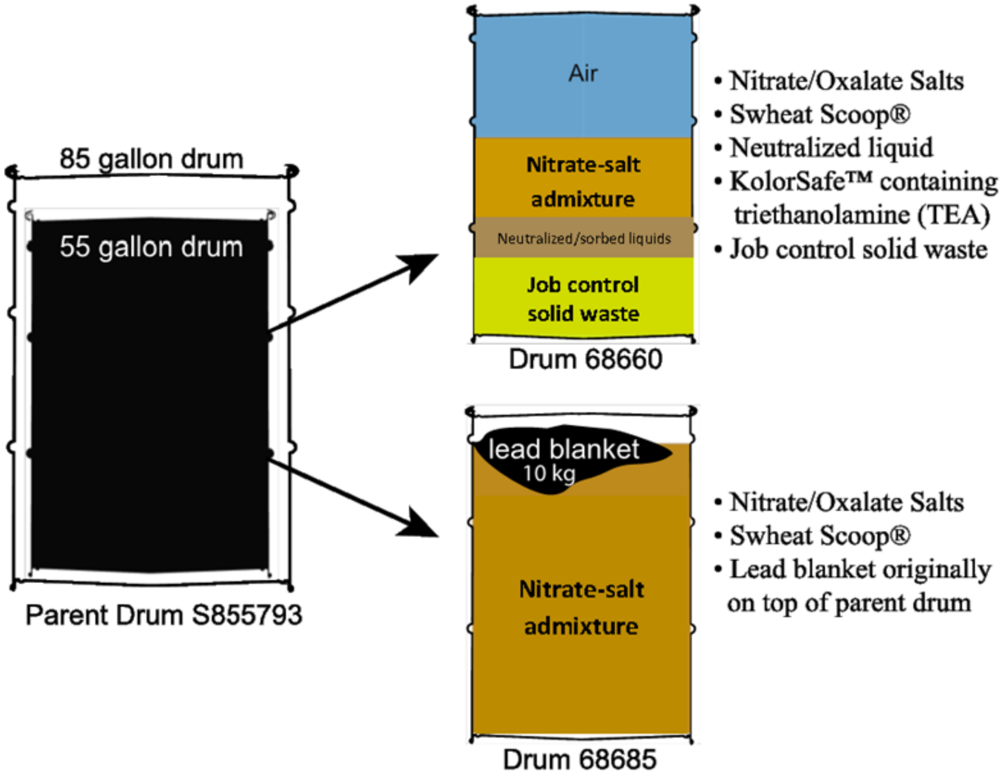

Figure E-1. Processing of parent Drum S855793 to produce sibling Drums 68660 and 68685

Figures E-2 and E-3 show the timeline for Drum 68660. Figure E-2 presents the data in flowchart form. Figure E-3 shows the actual timeline. Drum 68660 was produced December 4, 2013. It remained at Los Alamos until January 2014 when it was shipped to WIPP (January 28-29). It was placed in Room 7 Panel 7 two days later on January 31. On February 14, 2014, Drum 68660 breached and resulted in a radiation release.

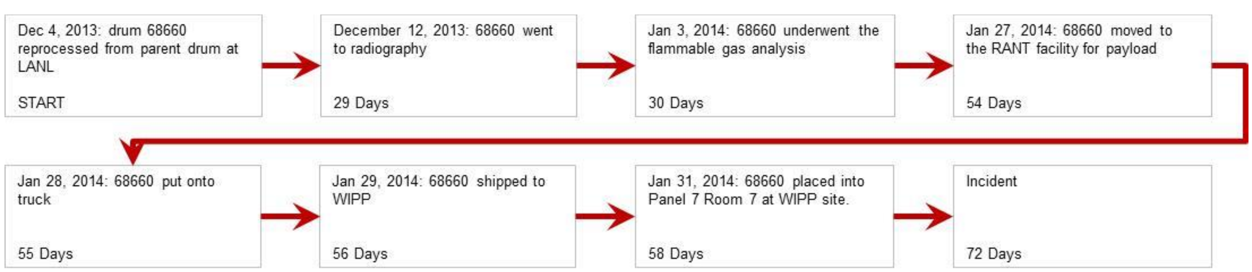

Figure E-2. Flowchart showing processing of Drum 68660

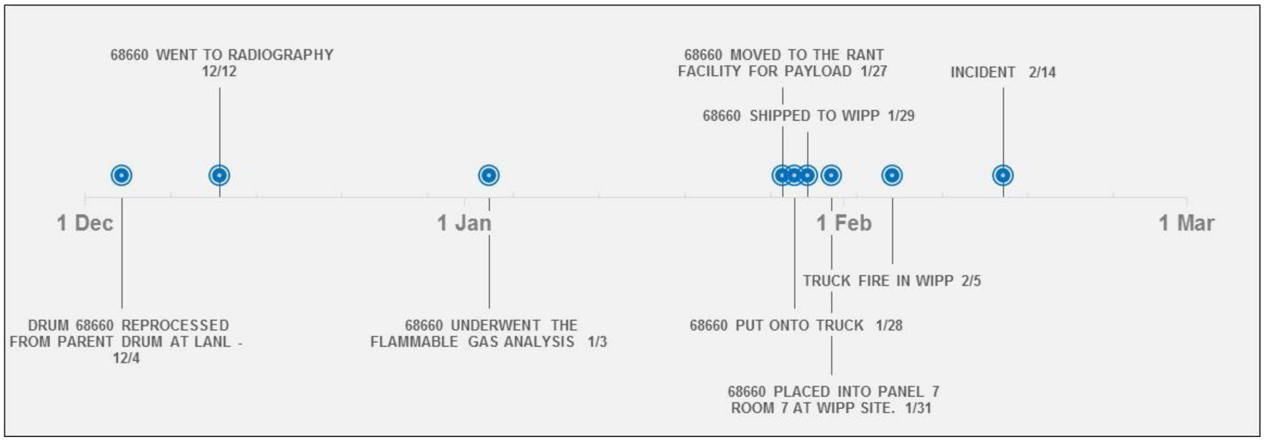

Figure E-3. Timeline for Drum 68660 from inception to the release event.

1.4 Drum Contents

Figure E-4 shows the assumed contents of Drum 68660. The yellow job control solid waste layer contains 5 kg of rubber and 6 kg of plastics. This layer also contains at least one glove of unknown composition (assumed to be tungsten-lined based on LANL information). The volume of the job control solid waste layer is 0.0403 m3 (13 gallons) and has a density of 273 kgm-3. This layer is assumed to be nonreactive in the finite element model. The brown “Swheat Scoop®” layer is subdivided into two layers: 1) the processed liquid layer and the 2) nitrate salt layer. The detailed content of the drum was assessed by performing a mass balance for the parent (S855793) and both sibling drums (68660 & 68685). The major assumptions used in this analysis are shown in the bottom of Figure E-4.

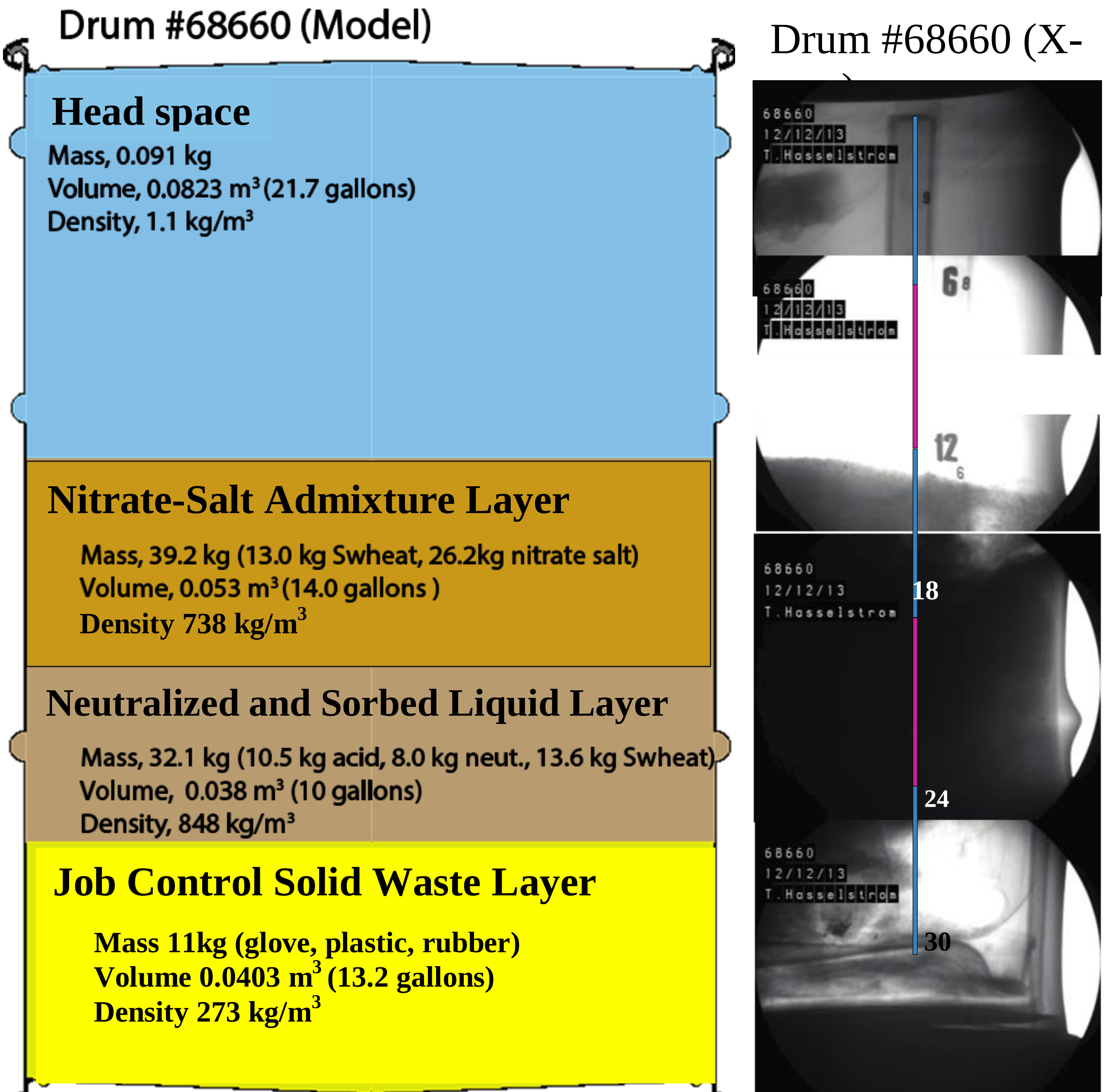

Figure E-4. TAT Model of Drum 68660 contents and distribution based on x-rays of Drum 68660

The content of Drum 68660 shown in Figure E-4 was determined with an overall material balance as given in Table E-1 using the assumptions listed in Table E-2. Table E-1 shows the net weight, which is the total mass minus the container mass composed of the drum steel, a plastic liner bag, and a fiberboard liner. The combined volumes of the processed layers were estimated from X-rays, which are partially shown in Figure E-4. The X-ray image in Figure E-4 was stitched together from an X-ray video and only shows the edge of Drum 68660. The combined processed salt and liquid layer volume probably ranges between 0.08-0.10 m^3 since the layer interfaces are not sharp. The Swheat Scoop® bulk density is higher than the optimal density mentioned in the Swheat Scoop® patent (550 kg/m^3) and is closer to 600 kg/m^3 based on measurements of large quantities of Swheat Scoop®. The higher density is due to settling in the larger volumes. The Swheat Scoop® moisture content is typically between 10-13% at the manufacturing facility and loses about 2% moisture when it is shipped to dry environments such as New Mexico. This would make the moisture content range between 8 to 11% on a mass basis. We have chosen 10% to be the moisture content in Swheat Scoop®. An overall mass balance was used to determine the total mass of Swheat Scoop® used to process the waste in the parent drum.

Two gallons of liquids were decanted from the nitrate salts in parent drum S855793. The two gallons of decanted liquids were composed of 3.3-molar nitric acid with dissolved nitrate salts. The two gallons of decanted liquids were neutralized with 2 gallons of 3.3-molar TEA making a total of 0.015 m^3 (4 gallons) of liquid that was absorbed using Swheat Scoop®. The *Swheat Scoop® to liquid* volume ratio was assumed to be 3:1 based on experience in our laboratory where large amounts of Swheat Scoop® were required to fully absorb liquid. The remaining mass of Swheat Scoop® was mixed with the nitrate salts that were distributed between sibling drums 68660 and 68685 giving a *Swheat Scoop® to nitrate salt* volume ratio of 0.7:1. This ratio is lower than the specification of 1.2:1 for zeolite absorbents.

The 11 kg of nonreactive job control solid waste was added to the bottom of Drum 68660 and included a tungsten glove, at least one empty TEA bottle, and plastic bags. The "waste" above the job control solid waste layer in 68660 is subdivided into two reactive layers: a *processed liquid layer* directly above the job control solid waste layer and a *processed nitrate salt layer* above the processed liquid layer. The space above the processed nitrate salt layer is filled with air. The volume and density of each of these layers is given in Figure E-4. The estimated contents of the processed salt layer and processed liquid layer are given in Tables E-3 and E-4, respectively. Table E-3 has a detailed description of the composition of the nitrate salts. Weisbrod estimated the composition of the nitrate salts by starting with Veazey salt analysis. Weisbrod conducted a series of evaporation simulations using Stream Analyzer© from OLI Systems. Table E-3 also lists a simplified composition for the nitrate salts. Measured solubility data at our laboratory were used with this simplified composition to determine the amount of nitrate salts that were dissolved into the decanted liquid as shown in Table E-4.

Table E-1. Parent and sibling drum mass balance

Name	Weight, kg	
Parent (S855793)		
Nitrate salts	115.91	
Decant liquid (3.3 molar HNO3 with dissolved nitrate salts)	10.48	
lead blanket (on top of drum)	9.98	
68725 net weight	136.37	
External materials added to parent waste		
Neutralizer (3.3 molar triethanolamine)	7.97	
Room trash (tungsten glove, plastic bottle, etc.)	11.00	
Swheat (10% moisture)	71.34	
External material net weight	90.31	
Sibling (68660)		
Nitrate salts	26.16	Remediated salt layer
Swheat (3:1 Swheat to liquid ratio by volume)	13.63	
Decant liquid (3.3 molar HNO3 with dissolved nitrate salts)	10.48	Remediated liquid layer
Neutralizer (3.3 molar triethanolamine)	7.97	
Swheat (0.7:1 Swheat to salt ratio by volume)	13.03	
Room trash	11.00	Trash layer
68660 net weight	82.27	
Sibling (68685)		
Nitrate salts	89.75	
Swheat (0.7:1 Swheat to salt ratio by volume)	44.68	
Lead blanket	9.98	
68685 net weight	144.41	
Mass parent + Mass external materials	226.68	✓
Mass 68660 + Mass 68685	226.68	✓

Table E-2. Assumptions regarding Swheat Scoop® and processed liquid

Assumption	*Source*	*Notes*
Processed layer volumes in both sibling drums (68660 and 68685)	Estimate from X-rays	X-ray of 68660 and 68685.
Swheat bulk density	Swheat patent (5,690,051 on 11/25/97) and measurements	We measured 550 kg/m^3 in small samples and 650 g/m^3 in larger samples. 600 g/m^3 is assumed to be the nominal density
Swheat moisture	Charles Neece (Pet Care Systems representative)	As shipped moisture content is 10-13%. In dry climates, this drops by 1-2%. The moisture content in NM is assumed to be 8-11 wt%.
Swheat:salt and Swheat: liquid volume ratio	Mass Balance and assumed volume ratio for Swheat:liquid.	The overall Swheat mass was determined by a parent to sibling mass balance. The volume ratio for the Swheat:liquids was assumed to be 3:1. The Swheat:nitrate salt volume ratio was calculated to be 0.7:1.
Neutralizer volume and molarity	Estimate	X-ray pictures showed a one gallon plastic neutralizer container in the trash layer. Two gallons of neutralizer is a conservative estimate with molarity between 1-5 mols/L (3.3 mols/L used in the current work).
Molarity of nitric acid	Determined from amount of neutralizer.	Assumed to be the same as the neutralizer.
Nitrate salt composition	Kirk Weisbrod analysis	Determined from "stream analyzer" simulations assuming 1) analysis of source waste, 2) stream ratios stored in parent drum, and 3) amount of liquid retained with solid crystals.

Table E-3. Processed salts in 68660 [a]

Composition	Detailed Mass, kg	Simplified Mass, kg
Dry wheat in Swheat	11.72	11.72
H_2O in Swheat	1.30	1.30
H_2O (with trace elements)	0.05	
$Al(NO_3)_3 * 9H_20$	0.62	
$Ca(NO_3)2 * 4H_2O$	2.48	2.70
KNO_3	0.54	
$Mg(NO_3)2 * 6H_2O$	16.30	17.74
$NaNO_3$	3.58	3.89
$Ni(NO_3)2 * 6H_2O$	0.02	
$Pb(NO_3)_2$	0.00	
$(COOH)_2$	0.40	
$Cr(NO_3)_3 * 9H_2O$	0.03	
$Fe(NO_3)_3 * 9H_2O$	1.69	1.84
HNO_3	0.40	
NaF	0.05	
Total	39.19	39.19

[a]Volume of this layer is 0.053 m^3

Table E-4. Processed liquids in 68660[a,b]

Composition	Mass, kg
Dry wheat	12.26
H_2O in Swheat	1.36
H_2O in decant	3.13
HNO_3 in decant	1.57
Nitrate salts[b] in decant	
$Ca(NO_3)_2 * 4H_2O$	0.54
$Mg(NO_3)_2 * 6H_2O$	4.27
$NaNO_3$	0.54
$Fe(NO_3)_3 * 9H_2O$	0.43
TEA in neutralizer	3.73
H_2O in neutralizer	4.25
Total	32.08

[a]Remediated liquid layer volume is 0.038 m^3.
[b]Based on measured solubility with composition based on most common salts.

1.5 Salt and Radiological Content in 68660

From the evaluation for source term there was an inference that the 14 individual salt bags in parent drum S855793 were mixed and processed with Swheat Scoop® as one batch prior to placement into the two sibling containers. From this finding the salt content can be determined accurately using the historical analytical data for each of the salt bags and the estimated mass of salts modeled to be in 68660 as described above. Table E-5 contains the material types, net weight of salts, and Pu and Am analytical information for each of the bags of MIN02 salts stored in parent container S855793 based on historical records.

Table E-5. MIN02 Salts in S855793 (Daughters 68660 and 68685)

Salt ID	MT	net (Kg)	Pu239 (g)	Pu240 (g)	Am241 (g)
10LALR1W	53	5.94	3.23	0.35	0.49
10LR5W1	53	4.8	0.47	0.05	0.12
10LR5E1	53	9.71	1.56	0.17	0.48
10LR16W1	53	8.6	2.20	0.20	0.50
10LR16E1	53	7.4	0.59	0.05	0.31
10LR17W1	53	7	0.71	0.06	0.40
10LR17E1	53	7.2	0.33	0.03	0.21
10RR18E1	52	9.68	4.08	0.25	0.39
10OX11W1-1	52	9.96	7.71	0.49	0.37
10OX11W1-2	52	10.97	8.49	0.54	0.41
10LR18E1	52	7.5	1.10	0.07	0.13
10LR18W1	52	12.93	2.13	0.14	0.39
10OX21E1	52	9.6	1.24	0.08	0.14
10OX21W1	52	15.1	2.27	0.15	0.08
	Total	**126.4**	**36.12**	**2.63**	**4.41**

Nine bags of salts (71.08 Kg) were produced from lean residues anion exchange feeds (“W” designates washes and “E” designates effluents). One bag (9.68 Kg) from rich residue anion exchange feed. And, four bags (45.63 Kg) from the anion exchange plutonium effluent after oxalate precipitation. Mixing these materials then placing 26 kilograms of salts into 68660 as predicted by the WIPP TAT models would result in the following inventories of salt types and Pu and Am isotopes shown in Tables E-6 and E-7 below. Also included in Table E-3 is the heat generated in the salts from radiolytic decay for each of the isotopes based on the masses listed. The Decay Heat constants used were 1.9, 6.8, and 114 W/Kg for Pu239, Pu240, and Am241 respectively.

Table E-6. Inventory of Nitrate Salt Types in Drum 68660

LR Feed Salts (Kg)	**RR Feed Salts (Kg)**	**OX Precip. Feed Salts (Kg)**
14.6	2.0	9.4

Table E-7. Pu and Am Inventory and Radiolytic Heat Generation from the Salts in Drum 68660

Pu239 (g)	**Pu240 (g)**	**Am241 (g)**
7.43	0.54	0.91
Decay Heat (W)		
0.014	0.004	0.104

These inventory values are based on the data as measured at the time of generation of the parent. At the time of processing and generation of the daughter drums there was 2 gallons of free liquid recorded to be in the parent which separated from the wet vacuum dried salts. The number and identification of which salt bags this liquid separated from is unknown so it should be noted that the loss of liquid weight will have an impact on the weights listed for each of the salt types in Table E-5 and the Decay Heat estimates in Table E-6. Assuming 3.3M HNO_3 and a density of 1.12 g/mL, 2 gallons of liquid would weigh 8.5 Kg. This is 6.7% of the total salt mass and would therefore have to come from multiple bags of the salts as all but one contained salts weights less than this.

The 2 gallons of processed liquid mixed with Swheat Scoop® and bagged separately was placed into 68660. Shown in Table E-8 are the historical Am and Pu analysis of the liquids leftover after flash evaporation of the waste streams. These liquids were removed for disposal by cementation however the results would be analogous to the activities in the interstitial liquid present in the moisture content of the vacuum dried salts. Note that if liquids were to be observed within a salt bag during processing the operators would have classified it a “containerized” liquid. That liquid would have been drained from the bag and processed with Swheat Scoop® separately from the salts. No such comment was recorded in the records for the processing of S855793.

Table E-8. Am and Pu Activity Concentrations Analyzed in the Liquids Phases after Flash Evaporation

Liquid from Salt ID	**Pu239 (Ci/L)**	**Pu240 (Ci/L)**	**Am241 (Ci/L)**	**Pu+Am (Ci/L)**
10LALR1W	0.055	0.14	1.23	1.43
10LR5W1	0.009	0.02	0.35	0.39
10LR5E1	0.019	0.06	1.07	1.14
10LR16W1	-	-	-	-
10LR16E1	0.007	0.02	0.72	0.75
10LR17W1	0.006	0.02	0.64	0.66
10LR17E1	0.005	0.01	0.69	0.71
10RR18E1	0.032	0.14	0.64	0.81
10OX11W1-1	0.004	0.02	0.06	0.08
10OX11W1-2	-	-	-	-
10LR18E1	0.009	0.04	0.36	0.41
10LR18W1	0.010	0.04	0.49	0.54
10OX21E1	0.011	0.04	0.24	0.29
10OX21W1	0.012	0.05	0.11	0.17

Analytical information missing for two liquids (10OX11W1-2 is a duplicate of 10OX11W1-1). Missing data not considered to have a major effect on the evaluation for an estimate of total activity in the 2 gallons of processed liquid.

To create a working range of isotopic Pu and Am activities to consider for the processed liquid it was simply assumed first that all of the individually bagged salts contributed equally to the free liquid volume, then the seven salts with the highest level of interstitial liquid activities contributed equally to the free liquid volume, and lastly, for the low range, seven of the lowest activity liquids. For the liquids with missing activities it was assumed 10OX11W1-2 equaled that of 10OX11W1-1 and 10LR16W1 equaled that of 10LR5W1 which is reasonable as the first can be considered a duplicate sample based on its identifier and the other a feed stream similar to the known (LR wash), again based on the identifier. The ranges for Pu and Am mass inventory and the associated radiolytic heat generation are listed in Table E-9 below.

Table E-9. Ranges for Pu and Am Inventory and Radiolytic Heat Generation in Drum 68660 Processed Liquid

All Salts Contributed Equally		
Pu239 (g)	**Pu240 (g)**	**Am241 (g)**
5.92	0.48	1.18
Decay Heat (W)		
0.011	0.004	0.134
7 Highest Activity Salts Contributed Equally		
Pu239 (g)	**Pu240 (g)**	**Am241 (g)**
7.94	0.66	1.84
Decay Heat (W)		
0.015	0.005	0.209
7 Lowest Activity Salts Contributed Equally		
Pu239 (g)	**Pu240 (g)**	**Am241 (g)**
3.91	0.29	0.51
Decay Heat (W)		
0.007	0.002	0.059

Comparison of Attributes of 68660 to Other Drums in Waste Room 7 Panel 7

There are 55 LANL storage drums containing MIN02 waste salts in Waste Room 7 Panel 7. Using WCRRF processing packages and Real Time Radiographs a comprehensive listing of physical items and attributes for all LANL MIN02 drums in Waste Room 7 Panel 7 was made. Below is a listing of key attributes associated with 68660 compared to other LANL drums in the room.

- 21 drums contain processed liquid. The processed liquid in 68660 had a recorded pH of 0. Five other drums with processed liquids have recorded pH’s of 3. The other 15 drums with processed liquid have no pH value recorded in the WCRRF paperwork.
- 3 drums contain one or more tungsten/bismuth/lanthanum-impregnated glovebox gloves and processed liquids. Review of the RTR of Drum 68660 performed at LANL indicated that the glovebox glove was in the job control solid waste layer at the bottom of the drum physically separated from the neutralized liquid/Swheat Scoop® layer.[1]
- 24 drums are from processed parents with some salts generated using oxalate precipitation. 11 of these drums have processed liquids (of these only 68660 contained a glovebox glove). Two drums in the room have an indeterminate salt makeup due to missing WCRRF processing packages.
- 9 drums were 60% or less in fill volume (5 of these also contained processed liquid)
- 11 drums resulted from the processing of parents with MT53 MT54 salt types

Below is a series of tables listing combinations of key attributes compared to 68660.

[1] See Appendix E, Addendum A, “Expert Interpretation Of Real Time Radiograph Recordings,” for details.

Table E-10. Drums with processed liquid in them and whose parents contained oxalate salts

Drum	**Location**	**Oxalates Salts in Parent (~ 2 wt%)**	**Processed Liquid**	**Glovebox gloves**	**Drum % Full**	**Parent Initial PH**
68545	13:3:B	Y (857 grams)	Y (5 gal)	N	100%	NR
68555	15:5:M	Y (1273 grams)	Y (7 gal)	N	60%	NR
68616	12:6:T	Y (990 grams)	Y (2 gal)	N	80%	NR
68629	5:1:T	Y (236 grams)	Y (1.1 gal)	N	90%	3
68654	5:1:T	Y (236 grams)	Y (8 gal)	N	90%	3
68655	5:1:T	Y (1073 grams)	Y (4 gal)	N	80%	NR
68660	16:4:T	Y (913 grams)	Y (2 gal)	Y	60%	0
68667	15:5:B	Y (832 grams)	Y (6 gal)	N	40%	NR
68672	3:3:T	Y (177 grams)	Y (3 gal)	N	80%	NR
68680	10:2:T	Y (572 grams)	Y (3 gal)	N	100%	NR
68687	15:5:B	Y (695 grams)	Y (3 gal)	N	100%	NR

Table E-11. Drums with processed liquids and glovebox gloves and whose parents contained oxalate salts

Drum	**Location**	**Oxalates Salts in Parent (~ 2 wt%)**	**Processed Liquid**	**Glovebox gloves**	**Drum % Full**	**Parent Initial PH**
68660	16:4:T	Y (913 grams)	Y (2 gal)	Y	60%	0

Table E-12. Drums with glovebox gloves

Drum	**Location**	**Oxalates Salts in Parent (~ 2 wt%)**	**Processed Liquid**	**Glovebox gloves**	**Drum % Full**	**Parent Initial PH**
68573	10:4:T	N	Y (7 gal)	Y	75%	NR
68660	16:4:T	Y (913 grams)	Y (2 gal)	Y	60%	0
68668	15:5:B	N	Y (3 gal)	Y	80%	3

Table E-13. Drums 60% full or less

Drum	**Location**	**Oxalates Salts in Parent (~ 2 wt%)**	**Processed Liquid**	**Glovebox gloves**	**Drum % Full**	**Parent Initial PH**
68512	10:6:B	N	N	N	50%	NA
68555	15:5:M	Y (1273 grams)	Y (7 gal)	N	60%	NR
68607	16:4:T	N	Y (0.12 gal)	N (RTR)	50%	NR
68609	13:3:B	Y (1273 grams)	N	N	40%	NA
68626	13:3:M	N	N	N	40%	NA
68649	15:5:M	N	Y (10 gal)	N	25%	NR
68660	16:4:T	Y (913 grams)	Y (2 gal)	Y	60%	0
68667	15:5:B	Y (832 grams)	Y (6 gal)	N	40%	NR
94152	15:5:B	N	N	N	50%	NA

2. SOURCE TERM

Drum 68660 was the source of the released radiological material.

2.1 Summary Conclusion

Based on the available photographic and video information, there is no evidence of involvement of any drum other than Drum 68660. This observation is supported by data from a series of non-destructive measurements made of the drum contents for acceptance into the WIPP facility and destructive analytical measurements obtained from a variety of sample locations in the facility, which includes debris from 15-5 and 16-4, surface smears, and constant air monitor (CAM) filters within WIPP as well as sampling of the Station A fixed air sampler (FAS) high efficiency particulate air (HEPA) filter. When this data set was compared to historical analytical data for the Pu and Am isotopic content for the processed wastes of the LANL drums stored in P7R7, it was determined to be consistent with a release from Drum 68660. Although when accounting for the analytical uncertainties inherent in the isotopic measurements, it cannot be concluded that no other drum contributed to the isotopic signature, from these data it can be stated that the dominant source of radioactivity was Drum 68660. Therefore, based on the evidence—photography and video; parent drum historical Uranium (U), Pu, and Am data for all LANL material types stored in P7R7; Central Characterization Program (CCP) gamma spectra for key LANL drums in P7R7 using high efficiency neutron counters (HENCs); and U, Pu, and Am isotopic measurements on post-event samples—it has been concluded that Drum 68660 was the source for the post-event radioactive contamination at the WIPP facility.

2.2 LANL Material Types and Panel 7 Drums' Salt Content

Visual evidence collected by the AIB positively identified a breached storage drum in Waste Room 7 of Panel 7 later identified as a LANL waste container. From storage records the container number was determined to be 68660. Using information documented in the Waste Characterization, Reduction, and Repackaging Facility (WCRRF) processing waste package it is recorded that 68660 and its sibling, 68685 (currently stored on the LANL reservation), were generated from the processing of parent waste container S855793 packaged in 1985 with 14 individual bags of legacy salts produced from the processing of LANL process waste streams consisting of plutonium material types (MT) 52 and 53. Early post-event sampling and analysis detected Pu240 content in the range of 7 weight percent or higher and Am241/Pu239 activity ratios from 15 to 26 from a number of remote sampling points. The concentration of Pu240 detected is consistent with LANL MT52 and MT53 or mixtures of those. Additionally, from discussions with LANL experts based on process knowledge and historical analytical data, it can be stated that MT53 and MT54 are more likely to contain higher levels of Am241 than those of MT42 and MT52 resulting in higher Am241/Pu239 activity ratios in those material types. Based on these very early analyses of key signature actinide isotopic ratios performed by the WIPP onsite laboratory and LANL analytical lab a release of radiological material from 68660 was confirmed. However, the WIPP TAT wanted to conduct a more exhaustive study that included all waste items in Panel 7 to determine whether 68660 was the sole contributor of the radiological material distributed throughout the facility for the final conclusion of source term.

All LANL material types (MT) contributing to the waste stored in Waste Room 7 Panel 7 with general descriptors for U, Pu, and Am content are shown in Table E-14 below.

Table E-14. Material Types (grams)

MT Code	Isotopic Description
12	Depleted U
42	> 60% Pu-242
51	< 4.00% Pu-240
52	4.00 < 7.00% Pu-240
53	7.00 < 10.00% Pu-240
54	10.00 < 13.00% Pu-240
81	Natural U

From this information it can be seen that a clear distinction of plutonium material types can be made based on the isotopic ranges shown.

In conjunction with this information a detailed listing of salt content for every LANL waste drum in Waste Room 7 was developed from LANL WCRRF waste packages and WIPP facility information for storage locations within the room. The information recorded in the WCRRF processing packages link each of daughter drums to a parent drum and provides specific data for each of the individual salt bags contained therein including net weight of the salts, material type, and grams of Special Nuclear Material (SNM). SNM consists of the weight sum for Pu239 and Pu240. Combined, these pieces of information were used to evaluate each item's key actinide isotopic signatures and to guide the sample isotopic analysis for data relating to source term in an effort to determine whether 68660 could be the sole contributor to the event as the early visual observations and analytical results would indicate.

Table E-15 contains the detailed listing with location of all LANL processed drums in Waste Room 7 Panel 7. Shown are the daughter and parent identifiers, the number of salt bags contained within the parent broken out by material type, and the percentage of SNM from material types other than MT52.

Table E-15. Processed LANL Waste Drums in Panel 7

Daughter	Parent	Bags: MT52:MT53:MT54	%SNM from MT other than 52
Location: R2C6			
68494	S910170 (missing WCRR package)	-	-
68652	S852590	10:0:0	0%
Location: R3C1			
68614	S864662	12:1:0	5%
68623	S870381	11:0:0	0%
68671	S863789	10:0:0	0%
Location: R3C3			
68571	S846055	8:0:0	0%
68635	S832499	6:1:0 (plus one bag MT42)	15%
68636	S832499	6:1:0 (plus one bag MT42	15%
68672	S853279	11:0:0	0%
Location: R5C1			
68541	S813676	4:0:0 (plus one bag MT42)	2%
68605	S822952	2:0:0 (plus one bag MT42)	34%
68629	S853326	9:0:0	0%

Daughter	Parent	Bags: MT52:MT53:MT54	%SNM from MT other than 52
68654	S853326	9:0:0	0%
Location: R5C1, continued			
68655	S852590	10:0:0	0%
Location: R10C2			
68501	S910170 (missing WCRR package)	-	-
68669	S853492	12:0:0	0%
68680	S863789	10:0:0	0%
Location: R10C4			
68573	S822952	2:0:0 (plus one bag MT42)	34%
68578	S816837	4:0:0	0%
68647	S844689	6:3:0	22%
Location: R10C6			
68394	S841320 (missing WCRR package)	-	-
68395	S825902	4:0:0	0%
68422	S824208	2:0:0	0%
68423	S824208	2:0:0	0%
68424	S824208	2:0:0	0%
68510	S846055	8:0:0	0%
68511	S861975	9:0:0	0%
68512	S825902	4:0:0	0%
68513	S852883	10:0:0	0%
68577	S842181	3:0:0	0%
68582	S833481	8:0:2 (plus one bag MT51)	0.6%
68618	S825810	3:0:0	0%
Location: R12C6			
68616	S853771	12:0:0	0%
Location: R13C3			
68545	S846088	0:6:4 (plus 5 bags MT81 and 4 MT12)	100%
68548	S846088	0:6:4 (plus 5 bags MT81 and 4 MT12)	100%
68576	S825730	2:0:0	0%
68581	S816692	5:0:1	6%
68609	S852895	10:1:0	25%
68626	S832144	5:0:0	0%
68653	S833846	6:3:2	42%
68659	S853279	11:0:0	0%
68666	S845072	6:3:1	69%
Location: R15C5			
68328	S891513	13:0:0	0%
68459	S823004	4:0:0	0%

Daughter	Parent	Bags: MT52:MT53:MT54	%SNM from MT other than 52
68555	S852895	10:1:0	25%
68649	S844689	6:3:0	22%
Location: R15C5, continued			
68667	S853492	12:0:0	0%
68668	S832150	5:0:0 (plus one bag MT42)	7%
68687	S870065	10:0:0	0%
94152	S842528	5:0:0	0%
Location: R16C4			
68333	S846107	8:0:0	0%
68607	S822952	2:0:0 (plus one bag MT42)	34%
68630	S818449	4:0:0	0%
68660	S855793	7:7:0	26%
68670	S832150	5:0:0 (plus one bag MT42)	7%

LANL Historical Data for the Waste Salts

In addition to the information presented in Tables E-14 and E-15 historical data for Pu239, Pu240, and Am241 isotopes were obtained from LANL archived analytical records for the numerous bags of salt wastes contained in the parent drums that were processed. These data were obtained for the WIPP TAT through Kirk Veirs, a staff chemist at LANL. The pertinent data for these isotopes and drums of interest will be presented and discussed as arguments are made for groupings of drums for possible contribution to the radiological release. It should be noted that for this evaluation it was calculated that the radioactive decay of Pu241 to Am241 has minimal impact to the Am241/Pu239 activity ratios when comparing historical analytical records dating back to the 1980's to post-event sample analyses.

Unfortunately historical data and the WCRRF processing waste packages for S910170 and S841320, parent drums of 68494 and 68501, and 68394, respectively, could not be found in the archived records. One of those, 68494, can be ruled out as a contributor to the radiological release based on visual observations as discussed later. For 68501 and 68394 only the raw gamma spectra of the drums performed using a High Efficiency passive Neutron Counter (HENC) at the Central Characterization Project (CCP) in conjunction with the Fixed Energy Response Function Analysis with Multiple Efficiency (FRAM) software package analyses, as discussed below, was used for this source term evaluation.

FRAM Am241/Am243 Ratios and Post Event Pu and Am Isotopic Analytical Measurements

In LANL's July 2014 report "*Waste Isolation Pilot Plant Radiological Release: Phase I Report*" it was reported that the FRAM software code, developed at LANL to analyze pulse height spectra generated by high resolution gamma detectors, was used to extract Am241/Am243 mass ratios from raw gamma spectra data obtained by the CCP during handling and transport processes of all waste drums before emplacement in the WIPP facility. During LANL's Phase I investigation into the event the FRAM analysis of the HENC gamma spectra for 68660 and four other drums packaged in the same seven-pack storage platform was performed in an effort to determine whether the drum's radiological content would yield a unique signature for the gamma emitting isotopes and distinguish it from the others drums on the platform which contained salts solely from MT52. From this it was determined that a unique Am241/Am243 signature existed for 68660 when compared to those four MT52 drums packaged in the same storage platform placed in Column 16, Row 4. Inquiries into this revealed that in some instances to produce plutonium material meeting MT53 standards MT56 would be added. MT56 has a uniquely high

Am243 concentration compared to other material types. This information was very useful for this study and from it, it was decided that having a measured ratio Am241/Am243 is highly desirable for source term. The WIPP TAT requested and obtained CCP gamma spectra for all waste drums in Waste Room 7 Panel 7 whose parent containers had processed waste salts from types MT53 and MT54 and as noted above for the daughter drums resulting from the processing of parent drums whose WCRRF packages and historical analytical data could not be found. These spectra were independently evaluated by both LANL and ORNL experts. The results of this evaluation are shown in Table E-16 below with the calculated average and uncertainties (2-sigma).

Table E-16. Waste Drum Am241/Am243 Content Based on FRAM Using CCP Gamma Spectra

Daughter	Location	Parent	FRAM (ORNL) Am241/Am243 (mass)	FRAM (ORNL) SD 2-sigma	FRAM (LANL) Am241/Am243 (mass)	FRAM (LANL) SD 2-sigma	Average Am241/Am243 (mass)	Average SD 2-sigma
Drums from parents containing MT52 plus MT53 and/or MT54								
68660	R16C4	S855793	482	66	476	62	479	90
68685	LANL		436	52	455	56	445	76
68581	R13C3	S816692	89984	55966	84706	40818	87345	69270
68653	R13C3	S833846	3836	608	4387	648	4111	889
68635	R3C3	S832499	969	102	1012	104	990	146
68636	R3C3		1188	174	1143	159	1166	236
68555	R15C5	S852895	318	76	229	63	274	99
68609	R13C3		283	74	260	73	272	104
68614	R3C1	S864662	682	249	667	206	675	323
68647	R10C4	S844689	1001	54	999	67	1000	86
68649	R15C5		1249	203	980	168	1114	264
68666	R13C3	S845072	1400	103	1402	116	1401	155
Drums from parents drums S841320 and S9100170 whose WCRR packages are missing								
68494	R2C6	S910170	302	27	318	29	310	40
68501	R10C2		298	17	325	28	312	33
68394	R10C6	S841320	3325	523	3159	605	3242	833
Drums from parents containing MT52 on seven-pack with 68660								
68333	R16C4	S846107	1029	77	1057	204	1043	218
68607	R16C4	S822952	18810	7655	15401	11132	17105	13510
68630	R16C4	S818449	47255	13659	42527	14540	44891	19949
68670	R16C4	S832150	77208	32492	85825	35650	81516	48235

From these results it can be seen that the drums have a measurably different Am241/Am243 mass ratio compared to Drum 68660 and also statistically similar ratios amongst all sibling pairs. It can be inferred from these statistically similar results obtained on a sibling pair that salt bags processed from the parent containers were mixed as one batch before placement into the daughter drums as the Am ratio is not expected to be similar between each salt bag and especially so for MT52 and MT53 as was in parent container S855793 (7 bags of MT52 and 7 bags of MT53 salts). This conclusion is further supported by the comparison of two independent efficiency responses that can be generated using FRAM. Using a set of gamma energies resulting from the decay of either Pu239 or Am241 an efficiency curve can be generated with the software. When these two independently generated efficiency curves overlap it can infer homogeneity of the material measured. This was the case for all sibling pairs listed in Table E-16. This is an important finding because it was not evident from the Energy Solutions waste operator

interviews or information contained in the WCRRF waste packages how exactly the salts were split between sibling pairs. Note that results for these 4 sets of pairs do not infer it is known that similar operations were performed prior to distribution of other parent salts into daughter containers.
All WIPP TAT post-event sample analyses details and results are reported separately in the SRNL and PNNL analytical reports. Only the measured Am241/Pu239 activity ratios, Am241/Am243 mass ratios, and Pu240/Pu239 mass ratios used for this study of source term are reported in Table E-17 below.

Table E-17. SRNL and PNNL Post-Event Am and Pu Ratios

Sample Type	Sample	Am241/Pu239 (activity)	Am241/Am243 (mass)	Pu240/Pu239 (mass)
	SRNL			
Airborne	FAS	26.1	444	
	CAM Filter #2	17.4	523	0.0770
	CAM Filter #3	15.6	461	
	CAM Filter #4	15.8	505	
	CAM Filter #6	19.2	485	
	CAM Filter #7	17.9	495	0.0760
	CAM Filter #8	22.2	466	
	CAM Filter #9	12.5	477	
	CAM Filter #11	12.1	493	0.0720
Panel 7 Swipes	Room 6	10.2		
	Room 1	9.0		
Panel 7 debris	R15C5-replicate 1 debris	5.11	501	
	R15C5-replicate 2 debris	4.87	470	
	R15C5-replicate 3 debris	5.27	471	
	R15C5-replicate 4 debris	5.53	458	
	R15C5-replicate 5 debris	4.68	411	
	R15C5-replicate 6 debris	5.95	416	
	R15C5 (Total)	5.24	455	0.0752
	R-15 C-5 SWB #1	5.45	455	
	R-15 C-5 SWB #2	11.1	509	
	PNNL			
	R-15 C-5 SWB	3.17	534	
	R-16 C-4 LIP	3.65		0.0780
	14-0752c Smear 2 Upper Right	5.15		
	14-0752d Smear 3 Lower Left	1.95		
	14-0752k Velcro	2.73		
	14-0752fBottom Tape	1.94	467	
	14-0752g Top Tape	3.29	472	
	Smear 1 R16 upper left	2.68		
	Smear 2 R16 upper right	2.63		
	Smear 3 R16 bottom left	2.03		
	Smear 4 R16 bottom right	2.39		
	Velcro backing R-16	3.31		
	Subsample 14-0753 particulate vial	2.74		
	Residual particles R-16	3.09		0.0777

A cursory review of the data reveals that there is high variability for the Am241/Pu239 activity ratios. The general trend being that the lowest values are measured closest to the source and increases with distance from the source (R16 and R15, to room swipes, to CAMS, and to Station A FAS). As a note LANL and

Carlsbad analytical laboratory measurements on the FAS also detected an Am241/Pu239 activity ratio in the range of 26. This observed trend can be attributed to fractionation of the Am241 and Pu239 isotopes during the event and transport through the WIPP facility and out via the ventilation system. Given the different chemical and physical properties of the two elements the observed fractionation is to be expected. Another complicating factor to consider when using the Am241/Pu239 ratio for source term is the processing history for each of the individual produced salts. A salt's Am241 concentration is highly dependent on how the separation to recover plutonium was performed. When the plutonium material is processed using an anion exchange separation the liquid effluent contains most of the Am. When that liquid effluent is run through the TA-55 evaporators, it produces salt with high Am/Pu ratios. When an oxalate precipitation process is used Pu is precipitated with oxalic acid post ion exchange. The liquids filtered off of the oxalate precipitate are therefore very low in Am and when these liquids go through the evaporation process the resultant salts will be low in Am and therefore have low Am/Pu ratios. Being cognizant of which process was used to produce the waste salts being evaluated would have to be an important consideration if Am/Pu ratios are to be relied upon in the evaluation for source term.

Given the variability in the historical processing and post-event measured Am241/Pu239 ratios it was determined that although in a general sense the Am/Pu signature could be used to distinguish possible contributors to the event the Pu240/Pu239 and Am241/Am243 ratios are much more reliable signatures being the two dominate elements in the waste stream and the isotope pairs will behave chemically and physically similar negating any variability due to fractionation. Table E-18 lists the calculated simple averages of all the SRNL and PNNL measured Am and Pu ratios including the calculated 2-sigma deviation of the results. Also included are results bands with the low and high values shown for each based on this deviation at the 95% confidence level.

Table E-18. Calculated Average Pu and Am Ratios for all PNNL and SRNL Samples and their 2-sigma Uncertainties

Am241/Am243 (mass)	**Standard Deviation (2-sigma)**	**Pu240/Pu239 (mass)**	**Standard Deviation (2-sigma)**
475	62 (13%RSD)	0.0760	0.0044 (6%RSD)
low	high	low	high
412	537	0.0716	0.0804

From Table E-18, it can be seen that the variability in the data is acceptable for the measurement techniques used (gamma spectrometry for Am isotopes and quadrupole ICPMS for the Pu isotopes) and therefore, statistically, the samples can be attributed to a single source.

2.3 Systematic Evaluations and Discussions for Source Term

<u>Drums with only MT52 salts</u>

The first grouping of drums studied was that which were processed from parent drums with salts solely resulting from MT52 processing. A review of the historical analytical data revealed that all MT52 drums contained salts whose Pu240/Pu239 mass ratio was approximately 0.065 which is distinctly lower than the upper limit for Pu240 content for that material type (at 7 wt%). It is also measurably below any of the values analyzed on the post-event samples and the average listed in Table E-18 with a 2-sigma uncertainty. A review of the historical records also shows that the MT52 salts in this population of drums generally do have lower Am241 content and therefore lower Am241/Pu239 activity ratios compared to MT53 and MT54. Of course this cannot be said for every batch of MT52 salts in these drums due to the Am content being dependent on the factors described above. An example for the Pu ratio and generally low Am241 content in MT52 salts is given in Table E-19. These data were obtained from historical records for the individual salts in parent drum S842528. This drum was processed into drum 94152, location R15C5, and its sibling drum 94151 which was located in Panel 6. Shown in the table are the salt identifiers for each of the salt bags in the parent, the calculated Pu240 wt% and ratios for Pu240/Pu239 (mass) and Am241/Pu239 (activity) based on the historical analytical data for each of the salts. Some data are missing for IXFS995 but based on the recorded Pu240 wt% the data can be assumed to be similar to the other salts in the drum. The total values are those calculated assuming all of the salts were mixed to make one batch then split into the two sibling drums.

Table E-19. Analytical Pu and Am Isotopic Data for MT52 Salts in S842528

Salt ID	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
IXFS995	6.05		
994OX58	6.47	0.0692	1.04
990OX57	5.97	0.0635	1.05
IXFS988	6.02	0.0641	0.32
IXFS986	5.97	0.0635	0.17
	Total	**0.064**	**0.936**

From what has been described above and the data in Table E-19, it can reasonably be concluded that the Pu and Am results measured cannot be attributed to drums whose radiological content is exclusively from MT52. This excludes 32 drums in Panel 7 for source term.

<u>Drums with MT42 salts</u>
The second grouping of drums studied contained those that had been processed from parent drums with salts resulting from process of MT52 as well as MT42. MT42 has a very high Pu242 content at > 60 wt%. All SRNL and PNNL analysis for plutonium isotopics included measurements for Pu242. This isotope was not detected in any sample. There are eight LANL waste drums in Panel 7 which were processed from parents with MT42 salts. Those drums are listed in Table E-20 below which is a condensed listing of Table E-15 showing only daughter drums whose parents contained MT42 salts.

Table E-20. LANL Processed Drums in Panel 7 Whose Parent Drums Contained MT42 Salts

Daughter	Parent	Bags: MT52:MT53:MT54	%SNM from MT other than 52
Location: R3C3			
68635	S832499	6:1:0 (plus one bag MT42)	15%
68636	S832499	6:1:0 (plus one bag MT42	15%
Location: R5C1			
68541	S813676	4:0:0 (plus one bag MT42)	2%
68605	S822952	2:0:0 (plus one bag MT42)	34%
Location: R10C4			
68573	S822952	2:0:0 (plus one bag MT42)	34%
Location: R15C5			
68668	S832150	5:0:0 (plus one bag MT42)	7%
Location: R16C4			
68607	S822952	2:0:0 (plus one bag MT42)	34%
68670	S832150	5:0:0 (plus one bag MT42)	7%

Drums 68635, 68636, 68541, and 68605 are in locations in the panel for which AIB visual observations confirmed were not affected by the event. All of the MgO bags are observed to be intact in rows 2, 3, and 4 and there is no observable damage to any of the waste items. Based on these observations and the absence of Pu242 it was concluded that these particular drums were not contributors to the event. For the remaining drums in the table the MT42 SNM (Pu239 and Pu240) contribution is high enough that release of their material would have resulted in gross levels of Pu242 in the debris field, swipes and airborne monitors/filters. Therefore these last remaining drums are also determined to not have contributed to the event based on post-event sampling and analyses.

Drums with MT12 and MT81 uranium

Drums 68645 and 69648 located in Row 13 Column 3 were processed from the same parent drum, S846088, which contained MT53, MT54, MT12, and MT81 salts. A review of the analytical records for those salt bags reveal that all of the MT53 and MT54 items have Pu240 greater than 8.9 wt% and therefore couldn't be responsible for the isotopics measured. Secondly, the drum contents are mixed MT12 and MT81 which are depleted and natural uranium. A listing of the salt bags processed from the parent is shown in Table E-21 below. Some of the items identifying information is either illegible or missing however this has no bearing on the listed material types present in the parent container. From this information it can be seen that these two daughter drums have thousands of grams of uranium content mixed with the Pu. All post-event measurements for uranium have been at the trace levels with isotopics slightly enriched in U235. Based on this information it can be concluded these drums were not contributors to the event.

Table E-21. Material Types in Parent Drum S846088

Salt ID	Net Wt. (Kg)	MT
LCU118N101	5.95	81
LCU12-895	6.29	53
LCU12-895	6.29	12
LCU12-895	6.28	81
UOX9-95NE1	14.32	53
LCU13103NE	1.59	53
LCU13103NE	1.59	12
LCU13103NE	1.59	81
LCU14119NE	1.88	54
LCU14119NE	1.87	81
ILLEGIBLE	13.31	54
-	14.31	12
-	14.31	81
UOX7-555A	13.99	53
-	4.09	53
-	4.09	12
NAX8-71NE1	26.82	54
UOX8-71NW1	14.55	54
LCU1170NW1	5.95	53
LCU117ONW1	5.95	12
LCU1513NW	7.16	54
LCU15132NW	7.16	12

Drum with MT51 salts

One drum in the panel, 68582 located in Row 10 Column 6, had a bag of MT51 salts documented in the WCRRF package to have been processed from the parent. The SNM in these are < 4 wt% Pu240. It was concluded that this drum cannot be attributed to the release due to the fact that only 0.6% of its SNM content is from MT54 and MT51 and the MT51 plutonium would "dilute" the MT54 contribution. Therefore the Pu240/Pu239 signature would be indicative to MT52 and below what was measured.

Drums missing WCRR processing packages and historical analytical data

FRAM analysis was used for evaluation for source term for drums 68501 (R10C2) and 68394 (R10C6) In the absence of historical analytical information and WCRRF processing records. Shown in Table E-16 above drum 68394 was analyzed using FRAM to have an Am241/243 ratio much higher than any of the sampled locations in the Waste Room and can be excluded based on this. Drum 68501 has a lower analyzed ratio averaging 312 +/- 33. Its exclusion from the source term can be confidently made based on this ratio and its 2-sigma uncertainty but since it is closer to the post-event average measured Am ratio than 68394 and no other data is available for evaluation, the Pu240/Pu239 ratio was also included in ORNL's FRAM analysis for it and its two sibling drums 68426 (stored at WCS) and 68494. From this evaluation the Pu240/Pu239 mass ratios were analyzed to be 0.022 (68426), 0.043 (68494), and below detection limits (68501). For the two sibling drums the ratio in the salt would be indicative of MT51. Heterogeneity of the Pu and Am isotopes in drum 68426 was noted based on the comparison of independently produced Am and Pu efficiency responses as described earlier. The very low Pu ratios analyzed in the two siblings of 68501 and plutonium gammas below detection in 68501 indicates a low overall Pu content. This additional plutonium information further supports exclusion of drum 68501 from source term.

Remaining drums with MT53 and MT54 salts

Remaining are eight drums which are listed in Table E-22 with location identified and the combined %SNM contribution from MT53 and MT54. Based on the %SNM contribution and the historical Am and Pu measurements for the salts in the parent drums, these have isotopic content that could be attributed to the radiological release.

Table E-22. Remaining Eight Drums with MT52:MT53:MT54 Salt Mixtures

Daughter	Parent	Bags: MT52:MT53:MT54	%SNM from MT other than 52
Location: R10C4			
68647	S844689	6:3:0	22%
Location: R13C3			
68581	S816692	5:0:1	6%
68609	S852895	10:1:0	25%
68653	S833846	6:3:2	42%
68666	S845072	6:3:1	69%
Location: R15C5			
68555	S852895	10:1:0	25%
68649	S844689	6:3:0	22%
Location: R16C4			
68660	S855793	7:7:0	26%

For these remaining eight drums a more thorough evaluation of their isotopic content is needed for evaluation of source term. Tables E-23 through E-28 lists the historical Am and SNM isotopic analyses for the salts contained in each of the parents which produced these drums. The total values listed are those calculated assuming all of the salts were mixed to make one batch then split into the respective daughter drums during processing. As discussed earlier statistically similar FRAM evaluations for Am241/Am243 supports mixing of the salts prior to processing with Swheat Scoop® and placement into the siblings.

Table E-23. Analytical Am and Pu Isotopic Data for Salts in S844689 (Daughters 68647 and 68649)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
IXF1039	52	6.08	0.065	3.63
IXF1040	52	6.00	0.064	0.99
IXF1041	52	6.01	0.064	2.61
IXF1045	52	5.96	0.063	1.76
1038OX73NE	53	8.21	0.089	2.43
1038OX73NW	53	8.21	0.089	6.14
1042OX74NE	52	5.88	0.062	1.59
1042OX74NW	52	5.88	0.062	0.70
1043OX75NE	53	9.61	0.106	0.42
		Total	**0.070**	**1.88**

Table E-24. Analytical Am and Pu Isotopic Data for Salts in S816692 (Daughter 68581)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
IXFS412FB	52	6.00	0.064	17.30
IXFS413FI	52	6.00	0.064	27.81
IXFS414FI	54	12.00	0.136	54.72
IXFS415FI	52	6.00	0.064	38.49
IXFS416FI	52	6.00	0.064	15.44
IXFS417FI	52	6.00	0.064	5.72
		Total	**0.068**	**27.81**

Table E-25. Analytical Am and Pu Isotopic Data for Salts in S852895 (Daughters 68555 and 68609)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
OX180306NE	52	5.90	0.063	1.26
OX179305NW	52	6.10	0.065	2.40
LR308NW1	52	5.93	0.063	5.57
LR308NE1	52	5.94	0.063	8.03
DSL311NW1	52	6.21	0.066	3.70
OX181310NW	52	5.90	0.063	3.63
UOX181310NW	52	5.90	0.063	1.35
LR309NW1	52	5.99	0.064	6.83
LR309NE1	52	5.99	0.064	8.07
DSL311NE	52	6.21	0.066	3.98
UOX11313NW	53	9.64	0.107	3.07
		Total	**0.074**	**3.90**

Table E-26. Analytical Am and Pu Isotopic Data for Salts in S833846 (Daughter 68653)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
IXFS-811-FA	52	6.00	0.064	12.36
OXF812-FA	52	6.00	0.064	18.49
IXFS-815-FA	53	9.09	0.100	18.36
IXFS-816-FA	52	5.27	0.056	10.31
IXFS818-FA	52	6.00	0.064	29.77
IXFS-819-FA	54	11.06	0.124	43.92
IXFS-820-FA	54	12.00	missing	missing
821OX40FC	52	6.00	0.064	6.10
IXFS-823-FA	53	9.75	0.108	25.51
IXFS-825-FA	52	6.00	missing	missing
IXFS-827-FA	53	9.89	0.110	7.69
IXFS-829-FB	52	6.50	0.070	15.61
		Total	**0.084**	**18.67**

Information missing for one bag each of MT54 and MT52 salts. Weight of salts is 4.6 and 5.9 Kg respectively. Missing data not considered to have a major effect on source term evaluation.

Table E-27. Analytical Am and Pu Isotopic Data for Salts in S845072 (Daughter 68666)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
1077OX86FB	53	8.24	0.090	1.68
IXF1078	52	6.03	0.064	5.48
IXF1079	52	5.80	0.062	3.64
1080OX87FA	53	8.42	0.092	2.51
1081UOX-2	54	11.84	0.134	1.95
1082OX88	52	5.95	0.063	0.96
IXF1083	52	5.91	0.063	4.53
IXF1084NE	52	6.02	0.064	3.62
IXF1084NW	52	6.02	0.064	3.85
1085OX89SE1	53	9.53	0.105	1.90
		Total	**0.084**	**2.49**

Table E-28. Analytical Am and Pu Isotopic Data for Salts in S855793 (Daughter 68660)

Salt ID	MT	Pu-240 (wt%)	Pu-240/Pu239 (mass)	Am-241/Pu239 (activity)
10LALR1W	53	9.69	0.107	8.28
10LR5W1	53	9.69	0.107	13.70
10LR5E1	53	9.69	0.107	17.02
10LR16W1	53	8.14	0.089	12.41
10LR16E1	53	8.14	0.089	28.65
10LR17W1	53	8.14	0.089	30.76
10LR17E1	53	8.14	0.089	34.83
10RR18E1	52	5.82	0.062	5.25
10OX11W1-1	52	6.00	0.064	2.63
10OX11W1-2	52	6.00	0.064	2.63
10LR18E1	52	6.00	0.064	6.58
10LR18W1	52	6.00	0.064	9.97
10OX21E1	52	6.21	0.066	6.14
10OX21W1	52	6.21	0.066	1.97
		Total	**0.073**	**6.70**

To further define the contents for this population of drums the measured Am241/Am243 for each of the drums determined using FRAM software code (Table E-16) was used in conjunction with the calculated Pu ratio for the mixed salts and WIPP TAT Am and Pu analytical measurements. Table E-29 is a summary of these data.

Table E-29. Daughters' Mixed Salts Pu240/Pu239 and Am241/Pu239 Ratios and FRAM Am241/Am243 Ratios

Ratio	**Daughter**								**SRNL and PNNL Result Bands**
	68647	**68581***	**68609**	**68653**	**68666**	**68555***	**68649***	**68660***	
Mixed Salts Pu240/Pu239 (mass)	0.0700	0.0680	0.0740	0.0840	0.0840	0.0740	0.0700	0.0730	0.0716 - 0.0804
FRAM Am241/Am243 (mass)	1000	87345	272	4111	1401	274	1114	479	412 - 537
Mixed Salts Am241/Pu239 (activity)	1.88	27.81	3.90	18.67	2.49	3.90	1.88	6.70	1.94 – 26.1

*Drums containing processed liquid

During WIPP TAT discussions the presence of processed liquids has been highlighted as an attribute of interest in regards to reactivity of drum contents Because of these lines of inquiry the four drums in this grouping with processed liquids have been identified in the table.

From the data shown only Drum 68660 contains homogenized salts with both Am241/Am243 and Pu240/Pu239 mass ratios within the ranges of those measured in the post-event samples by SRNL and

PNNL. The next closest match is sibling drums 68609 and 68555. These drums along with one other sibling, 68549 emplaced in Panel 6, were produced from parent S852895. The parent contained 11 salt bags, 10 containing MT52 (126 Kg total) and 1 containing MT53 (14 Kg total) salts. The Pu240/Pu239 mass ratios in the MT52 salts are around 0.06 while the MT53 salts have a ratio of 0.107. What drives the mixed salts, inferred by FRAM, to have to ratio stated is that the MT53 SNM content makes up ~25% of the total therefore driving up the calculated ratio. The addition of a high confidence Am241/Am243 ratio was very important in this case for source term.

Based on this comparison and arguments put forth in this study, the release of radioactive contamination in WIPP can be attributed to Drum 68660.

APPENDIX E. ADDENDUM A. EXPERT INTERPRETATION OF REAL TIME RADIOGRAPHIC RECORDINGS

The following is an account by independent assessor Steve Tallman (Level III RTR, RTR SME, AMWTP-ITG-INL) of his full review of the Drum 68660 Real Time Radiograph (RTR) to determine whether the glove and neutralized liquid/Swheat Scoop® were in contact. Tallman's assessment is that the RTR shows a space between the glove and neutralized liquid/Swheat Scoop® in Drum 68660 but that a definitive statement that the glove and liquid waste were or were not in contact cannot be made based solely on the RTR.

Summary Review of LANL Containers by Steve Tallman– 11/19/2014

I completed a review of RTR Recordings for 11 LANL containers, and completed answering a set of questions on some specific and general comments about these containers at the request of DOE's Mr. Roger Claycomb:

1. How many gloves in 68660? There was one glove confirmed, could see the fingers and cuff.
2. Are gloves in direct contact with the waste? There was no evidence of a separate horsetail, which would provide a layer of plastic. There is some space between the S3000 solid waste and glove, in my opinion, there doesn't look like there is any contact.
3. Are gloves in bottom of 68660 separately bagged? No evidence of a horsetail, in my opinion it was indeterminate on whether the glove was bagged.
4. In looking at other drums, was it standard practice to bag things separately? In my review of the other containers listed below, there were some indications of horsetails in the waste, mostly in the levels that held the S3000 solid waste. There were indications that some of the solids had been packaged separately, because you can see the rounded corners of some of the individual packages, and some separation within the waste on some of the containers.
5. Look at 68685- compare and contrast with 68660 - waste stream looked similar. Drums were inspected on different RTR systems, which make the waste look different, but there is a mixed combination of debris and solids waste in both. A brief description of each container is provided below.

I reviewed RTR Recordings for a total of 11 containers. Quite a bit of time was spent on the recordings for containers 68660 and 68685, in looking for detailed information. The recordings for the other containers were reviewed to confirm the waste was similar, and to provide a brief description of the waste contents. In general, all of the containers appeared to be the same waste stream; S3000 solid waste with a mixture of debris waste present. The look and density of the waste appears to be consistent. A brief description of each container;

Container ID Brief description

68660 **-** The container has a liner, plastic bag with horsetail (rest of reviews, description shortened to just horsetail), a small amount of debris waste on top, PPE, then layers of solid waste. At bottom, one glove, finger/cuff observed, debris waste, rigid liner lid, large small mouthed poly bottle, small pieces of metal debris. One area of dense debris adjacent to glove, couldn't tell what it is, other than confirm it is debris, and one small horsetail seen, coming up the side of the container, in the solids waste

68685 (sibling to 68660) **-** This container was inspected on a higher energy system than the other containers. The container has a liner, horsetail, quite a bit of plastic waste on top, what looks like lead pieces folded/stacked, solids waste, and small debris pieces mixed in with solids. Some of the solids waste has individual horsetails noted, for indication of separate bags.

68501 - The container has a liner, horsetail, solid waste, that is individually bagged, smaller horsetails, shape of the packages, some small debris mixed in with solids, rigid liner lid in with waste.

68511 - The container has a different packaging configuration - a pipe over-pack component (POC) with packaging to hold POC in place, waste looked to be bulk loaded into POC, plastic debris, small bags of solid waste.

68541 - The container has a liner, horsetail, lead sheet on top, layers of solid waste, and a small amount of miscellaneous debris waste on bottom.

68494 - The container has pieces of lead sheet cut up, an area of solid waste, denser near the top of the waste, small amount of debris waste at bottom.

68555 - This container has a liner, horsetail, pieces of lead sheet, layer of solid waste, rigid liner lid buried in solid waste.

68573 - This container has a liner, horsetail, layers of solid waste, with small pieces of debris mixed in, individual bags of waste, based on shape and appearance, no small horsetails evident, rigid liner lid buried, debris waste on bottom, lead/plastic.

68605 - This container has a liner, horsetail, solids waste with small amount of debris mixed in.

68607 - This container has a liner, horsetail, lead sheet/pieces, plastic debris, and bags of solid waste, based on shape/appearance.

68668 - This container has a liner, horsetail, lead sheet, solid waste, lead debris, and rigid liner lid at bottom.

APPENDIX E. ADDENDUM B.

In addition to Drum 68660 the RTR's on file were used for the assessment of the following LANL waste drums to visually document their inventory and attributes.

For the two siblings 68494 and 68501 there was no WCRRF processing package available. The RTR was used to document drum contents from the processing of their parent S910170.

For drum 68541 and siblings 68573, 68605, and 68607, the WCRRF processing package recorded that all of these were packaged with processed liquid. In the comment section it was also written that a pair of glovebox gloves was included as additional waste material added to the daughters. However it was not documented into which drum the glovebox gloves were added to.

It was recorded that drum 68511 contained a "lead" glove but it was not clearly documented as being put into the drum.

By inventory of items and attributes, 68668 is the closest match to 68660. No oxalate salts though.

Lastly, 68555 and 68680 have approximately the same mix of Material Types, oxalate salts and adsorbed free liquids as 68660.

Drum ID	**RTR Operator Observations**	**Additional notes**
68501	Liner, Horsetail – inner bag Solids, plastic bag ~50% utilized Homogeneous solids Plastic lid Fiberboard liner plastic liner bag Bottom appears to be dry Plastic bags	No WCCR package
68494	10/28/13 Scrap lead – flat piece ~100% utilized Horsetail – plastic liner bag Homogeneous solids Plastic bags at bottom of drum Fiberboard liner plastic liner bag Bottom appears to be dry Able to verify only one horsetail at the top – single layer confinement	No WCCR package Lead is at the top of the drum
68494R	Liner & liner bag Horsetail at top Scrap lead Homogeneous solids Bottom of container Fiberboard liner, no lid, liner bag Appears to be dry ~95% utilized – one layer containment	Replicate scan of the drum.
68541	Hasselstrom	

Drum ID	RTR Operator Observations	Additional notes
	Liner – no lid, bag, horsetail of the bag Scrap lead Homogenous solids **Bottom of drum** Horsetail for plastic bag Fiberboard liner Another horsetail for plastic bag ~70% utilized Appears to have two layer confinement	
68573	Simmons Rigid liner with no lid Plastic liner bag & horsetail Homogenous solids Leaded rubber glove A bunch of pairs of leaded rubber gloves Fiberboard rigid liner No lid No liquid Whole bunch of leaded rubber gloves at the bottom ~75% utilized One layer confinement	Frame captures to show gloves
68605	Simmons Rigid liner – no lid Horsetail Homogeneous solids Plastic bag, horsetail, homogenous solids Fiberboard liner Bottom of drum, bottom of liner appear to be dry ~95% utilized, two layers confinement	
68607	Simmons Rigid liner no lead Plastic liner, horsetail Plastic containers, scrap lead, homogenous solids **Bottom of drum** Fiberboard liner, no lid Appear to be dry ~55% utilized, two layer confinement	Empty bottles & lead on top of salts
68511	Maestas Center pipe overpack Liner is vented Vent for POC Rigid liner & liner bag inside POC Liner sleeve inside POC Homogeneous solids Metal hardware (scrap metal)	

Drum ID	RTR Operator Observations	Additional notes
	Bottom of drum Plastic sheeting 90 mil liner Bottom appears to be dry ~50% utilized	
68668	Simmons Rigid liner, no lid Plastic liner bag with horsetail Scrap lead (flattened sheets) Homogenous solids Plastic bag, horsetail Plastic bags, horsetails **Bottom of drum** Leaded rubber gloves 90 mil liner lid Fiberboard liner Bottom of drum, bottom of liner appear to be dry ~70% utilized Two layers of confinement	Lead is near the top Operator repeated plastic bags and horsetails (indicating multiples) Gloves were at the very bottom of the drum (look like they may be in contact with salt) Noted pretty thick sheet of lead (leaded window?)
68555	Simmons Rigid liner, no lid Plastic liner bag with horsetail **Center of drum** Plastic bag, horsetails Scrap lid, homogenous solids, plastic bags **Bottom of drum** 90 mil plastic liner lid Plastic containers Bottom of drum, bottom of liner appear to be dry 50% utilized Two layers of confinement	
68680	Maestas Liner Horsetail plastic liner bag Plastic bags Homogenous solids Scrap lead **Bottom of drum** Scrap metal Plastic bags Plastic lid Fiberboard liner Bottom of drum, bottom of liner appear to be dry	
68685	Simmons Scrap lead	Lead is on top of the waste

Drum ID	RTR Operator Observations	Additional notes
	Plastic horsetail Homogenous solid Bit more scrap lead Plastic bags (plural) horsetail **Bottom of drum** Plastic bag Homogenous solid Fiberboard rigid liner – no lid Another horsetail for another plastic bag Bottom of drum, bottom of liner appear to be dry ~85% utilized Two layers of confinement Some scrap metal at top of drum	